WHITHER NONLINEAR ACOUSTICS?

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INTRODUCTION

If we include consideration of wave propagation in an ideal gas, we can trace the origin of theoretical nonlinear acoustics, at least as far back as Poisson's work in 1808 [1]. The first experiments in air were done 1.27 centuries later [2]. With liquids Fox and Wallace [3] tried to explain experimental results on sound attenuation with a correct, but somewhat inadequate nonlinear theory. Keck and Beyer [4] used the equations of hydrodynamics and showed that nonlinear considerations lead to the prediction of nonlinear distortion, something that had been observed in fluids by optical techniques [5] and showed that the nonlinear equation for wave propagation in fluids has the same form as that for an ideal gas. Different thermodynamical quantities appeared in the nonlinear equations, however. It is significant that Keck and Beyer were able to perceive the inherent similarity of the two descriptions without becoming confused by the dissimilarities of certain details of equations describing ideal gases compared with those describing liquids.

Now we come to observation of the nonlinear properties of solids by acoustical techniques. Already in 1660 Hooke had published work on stress-strain relations in solids. The fact that he chose in 1676 to present his results in his famous anagram CEIIINOSSSTTUU, which was unscrambled in 1678 to read UT TENSIO SIC VIS, means that Hooke himself was aware of nonlinearity, but after two years decided that it could be ignored. From that time up to the 1960's when third order elastic constants were introduced, nonlinearity was considered an aggravation to be avoided. In the 1960's considerable effort was put forth to derive nonlinear equations describing wave propagation in solids to a consistent approximation. Nowadays such an effort may appear trivial, but in 1960 it was far from trivial. The number of terms in the matrices was very great when one considered wave propagation in arbitrary directions. It was a tour de force to write down all terms in the equation for a preselected direction. Change the direction, and all the terms in the equation had to be rederived. Gradually it became clear that the nonlinear wave equations in the pure mode directions all had the same mathematical form, even though the number of terms was quite large. This was a simplification that allowed us to focus our attention on the origin of the nonlinearity.

The purpose of this discussion is to show how simply the equations now can be derived for crystalline solids. Comparison with equations for liquids and gases allows us to define nonlinearity parameters in a consistent way for solids, liquids, and gases. Evaluation of numerical magnitudes allows us to make some cross-checks and to begin correlating other physical properties with specific magnitudes of the nonlinearity

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parameters. At the appropriate point I will issue a warning about unwarranted assumptions for mixtures of two or more substances having different nonlinearity parameters.

THEORETICAL CONSIDERATIONS

The most direct derivations of the nonlinear wave equation is accomplished by defining independent variables

$$a_i,\ t \qquad (i = 1,2,3) \tag{1}$$

and dependent variables

$$x_i \qquad (i = 1,2,3), \tag{2}$$

the strain matrix

$$\eta = \frac{1}{2}(J^* J - I), \tag{3}$$

where the Jacobian matrix J has the elements

$$J_{k\ell} = \frac{\partial x_k}{\partial a_\ell}, \tag{4}$$

and the elastic potential energy

$$\Phi(\eta) = \frac{1}{2!}\sum_{ijk\ell} C_{ijk\ell}\eta_{ij}\eta_{k\ell} + \frac{1}{3!}\sum_{ijk\ell mn} C_{ijk\ell mn}\eta_{ij}\eta_{k\ell}\eta_{mn} + \cdots \tag{5}$$

Using $\Phi(\eta)$ in the Lagrangian function

$$L = \frac{1}{2}\sum_{i=1}^{3} \rho\left(\frac{\partial x_i}{\partial t}\right)^2 - \Phi(\eta) \tag{6}$$

allows us to use Langrange's equations

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{x}_i}\right) + \sum_{k+1}^{3} \frac{d}{da_k}\left(\frac{\partial L}{\partial\left(\frac{\partial xi}{\partial a_k}\right)}\right) = 0 \tag{7}$$

to derive the nonlinear wave equations. With the a_1 axis along the propagation direction the nonlinear wave equations become

$$\rho\ddot{x}_i = \frac{\partial}{2a_1}\sum_{K=1}^{3} J_{ik}\frac{\partial\Phi(\eta)}{\partial\eta_{1k}}. \tag{8}$$

This form of the nonlinear wave equations shows the central role played by the strain energy in determining the effect of medium nonlinearity. Choosing the x_k axes parallel to the a_k axes and defining the particle displacement $\xi_k = x_k - a_k$, one obtains the nonlinear wave equation in the form

$$\rho \frac{\partial^2 \xi i}{\partial t^2} = \sum_{j=1}^{3} A_{ij} \frac{\partial^2 \xi j}{\partial a_1^2} + \sum_{j,i=1}^{3} B_{ij\ell} \frac{\partial \xi j}{\partial a_1} \frac{\partial^2 \xi \ell}{\partial a_1^2} \tag{9}$$

which shows the mathematical form of the nonlinear equations required to describe the propagation of a wave in a nonlinear solid. Here the A_{ij} are linear combinations of second order elastic (SOE) constants which depend on the propagation direction and the $B_{ij\ell}$ are linear combinations of SOE constants and third order elastic (TOE) constants.

The same functional form as Eq. 9 is retained if one specializes to the principal directions in a cubic lattice. In such a case one can write the nonlinear wave equation in a form in which the linear combinations of SOE constants and TOE constants are explicitly separated. Let K_2 represent the linear combination of SOE constants and K_3 the linear combination of TOE constants. The nonlinear wave equation then takes the form

$$\rho_o \frac{\partial^2 \xi}{\partial t^2} = K_2 \frac{\partial^2 \xi}{\partial a^2} + (3K_2 + K_3) \frac{\partial \xi}{\partial a} \frac{\partial^2 \xi}{\partial a^2}, \tag{10}$$

and the combinations K_2 and K_3 for the principal directions in a cubic lattice are given in Table I. Writing the equations in this way emphasizes the fact that waves propagating in the principal directions in a crystalline lattice are described by nonlinear equations of the same mathematical form. The difference is in the magnitude of the coefficients.

It may be worthwhile to point out that the nonlinear equations describing wave propagation in crystalline solids are very similar in mathematical form to those describing waves in liquids and gases. This can be seen by beginning with the equations of Keck and Beyer [4]. By using an equation of state written in a Taylor series, Keck and Beyer showed that the nonlinear wave equation for fluids can be written

$$\frac{\partial^2 \xi}{\partial t^2} = C_0^2 \frac{\dfrac{\partial^2 \xi}{\partial a^2}}{1 + \left(\dfrac{\partial \xi}{\partial a}\right)^{B/A+2}}. \tag{11}$$

Specializing to ideal gases it becomes

Table 1. K_2 and K_3 for [100], [110], and [111] directions in a cubic lattice.

Direction	K_2	K_3
[100]	C_{11}	C_{111}
[110]	$\dfrac{C_{11} + C_{12} + 2C_{44}}{2}$	$\dfrac{C_{111} + 3C_{112} + 12C_{166}}{4}$
[111]	$\dfrac{C_{11} + 2C_{12} + 4C_{44}}{3}$	$\dfrac{C_{111} + 6C_{112} + 12C_{144} + 24C_{166} + 2C_{123} + 16C_{456}}{9}$

$$\frac{\partial^2 \xi}{\partial t^2} = C_0^2 \frac{\dfrac{\partial^2 \xi}{\partial a^2}}{1 + \left(\dfrac{\partial \xi}{\partial a}\right)^{\gamma+1}}. \tag{12}$$

They pointed out that B/A +2 for fluids in general plays the same role as ξ+1 for ideal gases. By writing a power series expansion of Eq. 11 we can expand on this point. Expanding the denominator in Eq. 11, one can write

$$\frac{\partial^2 \xi}{\partial t^2} = C_0^2 \left(\frac{\partial^2 \xi}{\partial a^2} - \left(\frac{B}{A} + 2\right) \frac{\partial \xi}{\partial a} \frac{\partial^2 \xi}{\partial a^2} + \ldots \right). \tag{13}$$

Comparing Eq. 13 with Eq. 10 reveals that B/A + 2 in Eq. 13 plays the same roles as $-\left(\frac{3K_2+K_3}{K_2}\right)$ in Eq. 10. One thus defines the nonlinearity parameter

$$\beta_s = -\left(\frac{K_3}{K_2} + 3\right) \qquad \text{for solids} \tag{14}$$

and

$$\beta_f = \frac{B}{A} + 2 \qquad \text{for fluids.} \tag{15}$$

Table II presents the results of such comparisons among gases, liquids and solids. Although this table results only from a comparison of the physical behavior of waves described by including nonlinear terms in the wave equation, a number of inferences can be made about physical nonlinearities, and some correlation of the magnitudes of the nonlinearities of the various media is possible, as is correlation of nonlinearities and other physical parameters. However, we first need to understand how one most readily evaluates the nonlinearity parameters of different media.

Table II. Parameters Entering into the Description of Waves of Finite Amplitude in Gases, Liquids, and Solids.

PARAMETER	IDEAL GAS	LIQUID	SOLID
C_0^2	$\frac{\gamma P_0}{\rho_0}$	A/ρ_0	K_2/ρ_0
Nonlinearity Parameter β	$\gamma + 1$	B/A +2	$-(K_3/K_2 + 3)$
B/A	$\gamma - 1$	B/A	$-(K_3/K_2 + 5)$
Discontinuity Distance L	$\frac{2C_0^2}{(\gamma+1)\omega U_0}$	$\frac{2C_0^2}{\left(\frac{B}{A}+2\right)\omega U_0}$	$\frac{2C_0^2}{-\left(\frac{K_3}{K_2}+3\right)\omega^2 \xi_0}$

MEASUREMENT OF NONLINEARITY PARAMETERS

Essentially two techniques exist for the evaluation of nonlinearity parameters: 1. Measurement of sound velocity as a function of applied stress, and 2. Measurement of harmonic generation in an initially sinusoidal ultrasonic wave. Because of space limitations we will describe only harmonic generation.

A perturbation solution of Eq. 10 gives the following results for longitudinal wave propagation in a fluid, an anisotropic solid, or along the principal directions in a cubic lattice:

$$\xi = A, \text{Sin}\,(ka\text{-}\omega t) - \left(\frac{K_3}{K_2} + 3\right)\frac{A_1^2 k^2 a}{8}\,\text{Cos}\,2(ka\text{-}\omega t) + \ldots \qquad (16)$$

With the solutions in this form one can interpret data from gases, liquids, and solids by appropriate use of Tables I and II and the definitions of nonlinearity parameters given in Eqs. 15 and 16. We will describe the procedure for solids since the harmonic generation technique has given most useful results in crystalline solids.

Equation 16 predicts that an initially sinusoidal wave generates its own harmonics as it propagates, and that the second harmonic grows linearly with distance and quadratically with amplitude and frequency. Measurements can be made with a capacitive microphone system described previously [6]. Samples are lapped optically flat with parallel surfaces in the direction of propagation of the ultrasonic wave. If necessary, correction can be made for diffraction [7]; however with samples as large as 1.5 cm on a side diffraction corrections can be negligible. Since parallel plate capacitive microphones can be calibrated absolutely, direct measurement of the amplitudes of the 30 Mhz fundamental and its second harmonic gives information sufficient to evaluate the harmonic generation described by Eq. 16. Plots of A_2 vs A_1^2 show that for most solids Eq. 16 is a valid description of initial harmonic growth. An example for silicon is given in Fig. 1. The linear behavior predicted by Eq. 16 is accurately followed in the [100], the [110], and the [111] directions in silicon. The slopes of these curves are direct measures of the nonlinearity parameters for the corresponding directions in silicon. Such behavior is found in most solids. Thus the harmonic generation technique is providing a wealth of information for interpreting the behavior of solids. Plots of the nonlinearity parameters of a number of crystalline solids of cubic symmetry present in Fig. 2 illustrates this point. Fig. 2 also shows that reliable information about nonlinearity parameters can be taken over a wide range of temperatures.

Since nonlinearity parameters are pure numbers, it might be useful to define the relative nonlinearity of crystalline solids and fluids. Table III lists nonlinearity parameters of various fluids. For the fluids given the nonlinearity parameters cover the range from 6 to 14. For comparison, we plot in Table IV values of the nonlinearity parameters of cubic solids for the [100] direction. They cover the range from 2 to 15. Thus the range of nonlinearity parameters of crystalline solids is wider than that of fluids. Further, there is a good correlation of the nonlinearity parameter with crystalline structure and bonding. The zincblende structure with covalent bonding generally has a small nonlinearity parameter. The anharmonicity of the potential function describing the motion of an atom in this structure has been used [8], in combination with the Keating model [9] to determine a complete set of six third order elastic constants of silicon and germanium. Only three harmonic generation measurements were used over the temperature range between 3 and 300° K. Many more details are available [10] about the correlation of the nonlinearity parameter in zincblende structure crystals with the interatomic potential functions. Over the entire range of parameters, shown in Table IV, Cantrell [11] points out that β depends exclusively on the Born-Mayer hardness parameter, which indicates that β is largely determined by the shape of the interatomic potential function. With the larger nonlinearity parameters one finds an increasingly important zero-point energy contribution. The

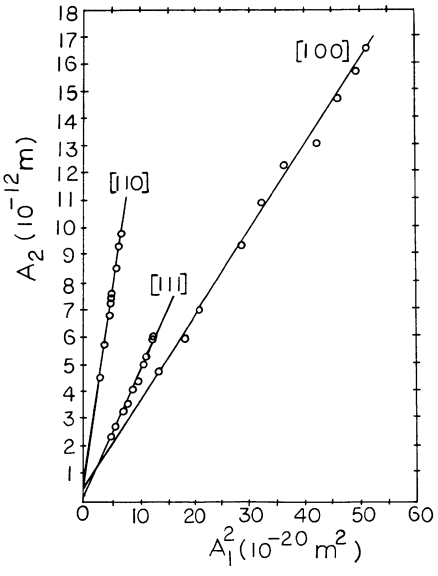

FIG.1 Measured second harmonic amplitude in silicon plotted as a function of the square of the fundamental amplitude.

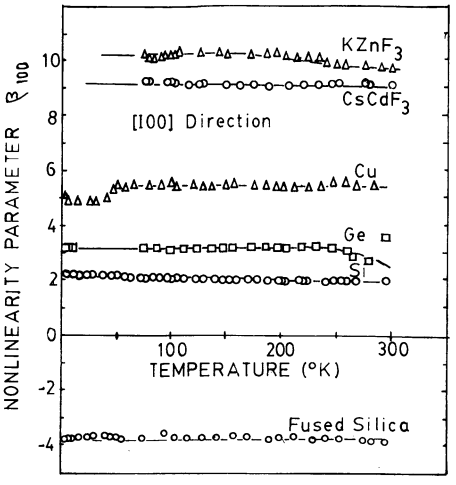

FIG. 2A Nonlinearity parameters in the [100] direction in cubic crystals between 3 and 300° K.

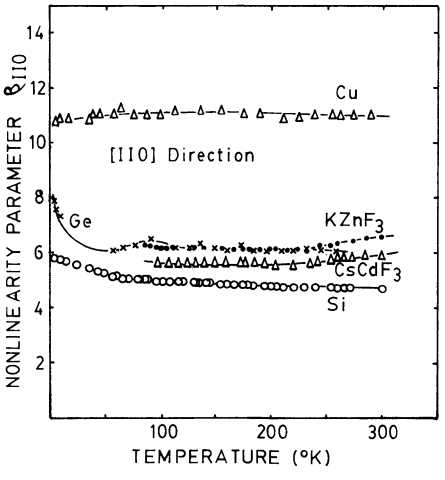

FIG. 2B Nonlinearity parameters in the [110] direction in cubic crystals between 3 and 300° K.

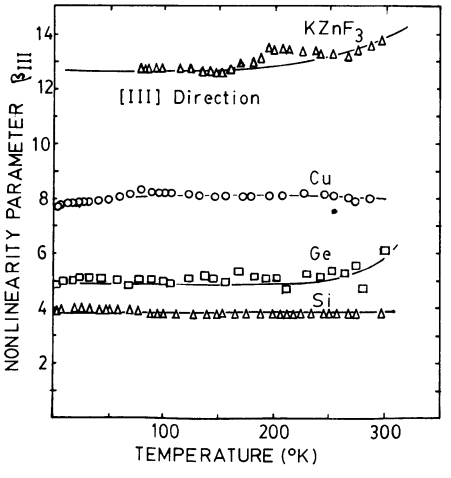

FIG. 2C Nonlinearity parameters in the [111] direction in cubic crystals between 3 and 300° K.

nonlinearity parameter also has been correlated with the Anderson-Grüensen parameter in solids [12] and temperature dependence of third order elastic constants of NaCl-structure alkalide halide crystals [13]. Of more practical technological interest is the fact that the nonlinearity parameter has been correlated with the ultimate yield strength of solids [14] and with hardness in steels [11].

Table III. Values of B/A and β for fluids at atmospheric pressure

LIQUID	TEMPERATURE (°C)	B/A	$\beta = B/A + 2$
Water-Distilled	0	4.16	6.16
	20	4.96	6.96
	40	5.38	7.38
	60	5.67	7.67
	80	5.96	7.96
	100	6.11	8.11
Acetone	30	9.44	11.44
Benzene	30	9.03	11.03
Benzyl Alcohol	30	10.19	12.19
CCl_4	30	11.54	13.54

Table IV. Comparison of Structure, Bonding, and Acoustic Nonlinearity Parameters Along the [100] Direction of Cubic Crystals [11].

STRUCTURE	BONDING	β_{AV}	RANGE OF β
Zincblende	Covalent	2.2	1.8- 3.0
Flourite	Ionic	3.8	3.4- 4.6
FCC	Metallic	5.6	4.0- 7.0
FCC (inert gas)	Van der Waals	6.4	5.8- 7.0
BCC	Metallic	8.2	7.4- 8.8
NaCl	Ionic	14.6	14.0-15.4

Nowadays correlations are made between nonlinear properties of solutions of biological substances, even tissue characterization of such inhomogeneous substances as muscle tissue [15], and in my laboratory, inhomogeneous solids such as $LiNbO_3$ ceramics and high T_c superconductors such as $YBa_2Cu_3O_{7-\delta}$. Although such correlations probably exist, one must be especially careful in taking the data required for their confirmation. The reason for caution is the fact that an implicit assumption has been made in the theoretical development that the substance is homogeneous, and hence that scattering of the ultrasonic wave does not take place. In inhomogeneous materials, both solids and liquids, scattering

most certainly does take place. The investigator therefore must be especially observant and accurately evaluate the effect of scattering on his data. Only then can truly meaningful nonlinearity parameters of inhomogeneous materials be reliably defined so that the measured magnitude of the nonlinearity parameter truly reflects the fundamental physical properties of the propagating medium.

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